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Nuclear magnetic relaxation of ^{133}Cs of distorted triangular antiferromagnet Cs_2CuBr_4

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Abstract

A quasi-two dimensional antiferromagnet Cs_2CuBr_4 is characterized as a frustrated spin system on a distorted triangular lattice. ^{133}Cs-NMR experiments on Cs_2CuBr_4 have been performed in the range of the magnetic field applied along b-axis up to 15.9 T, which covers the field range for the 1/3 magnetization plateau. Field dependence of T_1^{-1} at 0.5 K shows a hysteresis around each end of the plateau, indicating a field induced first order phase transition. We also present NMR spectrum indicating hysteresis and two phase coexistence around the transition field.

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1. Introduction

Recently, copper compounds with the magnetic Cu^{2+} ion which carry $S = \frac{1}{2}$ have been provided various interesting phenomena of quantum spin systems. A quasi-two dimensional antiferromagnet Cs_2CuBr_4, has attracted much interest, since the 1/3 plateau was observed in the magnetization curve in the field applied along the *b*- and *c*-axes [1]. Moreover, the 2/3 plateau was also found quite recently [2]. In the crystal of Cs_2CuBr_4, a distorted triangular lattice of Cu^{2+} ions is formed in the *bc*-plane. Then, these plateaux are considered to be stabilized by quantum fluctuation of $S = \frac{1}{2}$ triangular lattice antiferromagnet, which was discussed by theoretical and numerical studies [3–5]. Neutron diffraction experiments have suggested a cycloidal incommensurate spin ordering and a commensurate spin ordering for outside and inside of the plateau region, respectively, below $T_N \simeq 1.4$ K [2]. Therefore, when the field is increased below T_N, there are incommensurate-commensurate transitions at both ends of the plateau. It is also interesting that both the long-range ordering and the magnetization plateau occur simultaneously.

In order to study the spin dynamics and the spin structure in Cs_2CuBr_4 from a microscopic viewpoint, we have performed ^{133}Cs-NMR experiments in a temperature range down to 0.4 K and in a magnetic field range up to 15.9 T applied along *b*-axis which covers the 1/3 magnetization plateau region (approximately 13.9 ∼ 15.0 T for our sample).

We first discuss the experimental results of NMR spectrum and then turn to the relaxation rate.

2. Experimental method

We used the same sample of a single crystal Cs_2CuBr_4 throughout our measurements, because the critical fields of the 1/3 plateau slightly depend on the sample [6]. A conventional pulsed NMR method was used and ^{133}Cs-NMR spectrum was obtained as a function of the frequency under a constant magnetic field. As shown in our previous paper [7], two groups of NMR lines were observed in all cases, consistent with two inequivalent cesium sites, called as A and B sites, in the crystal of Cs_2CuBr_4. In this paper, we devote our attention to the results for B site.

In order to clarify the property of the phase transition, it is important to investigate whether there is a magnetic hysteresis or not. The hysteretic behavior was studied by the following procedure: For the measurement of increasing field, the applied field was increased monotonously step by step from a sufficiently low field; then, the data for decreasing field were obtained in the opposite way.

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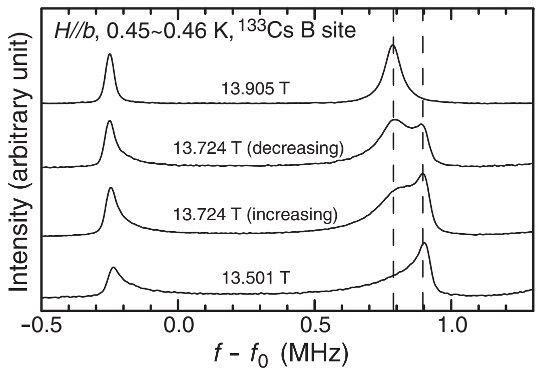

Fig. 1. NMR spectra of ^{133}Cs B site obtained at 0.45∼0.46 K under the field applied along b-axis. The dashed lines are guides for the eyes. The abscissa shows the shift in frequency. A field hysteresis is observed in the spectra at 13.724 T.

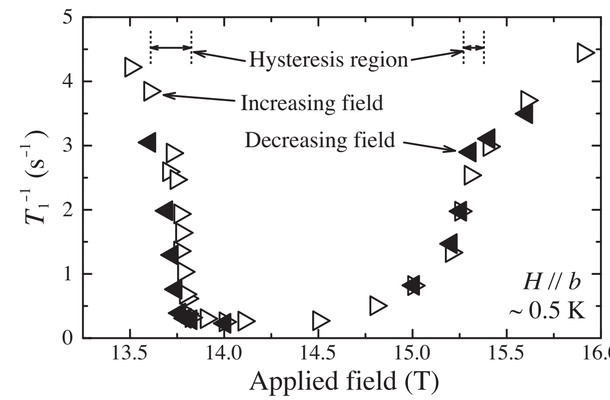

Fig. 2. Field dependence of T_1^{-1} at 0.5 K for the resonance line at lower frequency of B site. The open triangles and solid triangles indicate the data for increasing and decreasing field, respectively. The dotted lines indicate regions in which hysteresis of T_1^{-1} is observed.

3. Experimental results and discussion

Figure 1 shows NMR spectra of B site of ^{133}Cs measured at 0.45∼0.46 K under the external magnetic field applied along b-axis. The resonance line split into two peaks below T_N. The spectrum at 13.5 T below the plateau region shows a double-horn type line shape with a continuum of finite intensity between the two peaks. Such a line shape is a signature of an incommensurate spin structure. In the cycloidal phase, the hyperfine field caused at cesium nuclei should have a continuous distribution. The spectrum at 13.9 T is measured in the plateau region near the lower critical field. As the spectrum at 14.5 T about the center of the plateau shown in our previous paper [7], the continuum between the peaks disappears and the spectrum consists of a discrete two peaks for each site, which suggests the realization of up-up-down spin structure. This feature was observed at any position in the plateau region.

As shown in Fig. 1, the spectrum of 13.724 T in increasing field is clearly different from that at the same field of decreasing field. Furthermore, in each spectrum at 13.724 T, a double-peak structure appears around the upper peak of the resonance line of B site. These peaks appear at almost the same position as the upper peaks for commensurate (13.501 T) and incommensurate (13.905 T) phases, respectively. Therefore, the coexistence of the two phases is strongly suggested.

We found the hysteretic behavior at each end of the plateau also in a field dependence of the nuclear spin-lattice relaxation rate T_1^{-1} shown in Fig. 2. Here, T_1^{-1} is measured at 0.5 K for the lower frequency peak of the resonance line of B site which has an almost constant shift as seen in Fig. 1. These results of the spectrum and T_1^{-1} provide an evidence of first order phase transitions at both ends of the plateau region. It is also found that the change of the value of T_1^{-1} in the hysteresis around the upper critical field which was observed between 15.3 ∼ 15.4 T was rather small as compared with that around the lower critical field between 13.6 ∼ 13.82 T. This means that the two transitions are somewhat different from each other. Such a difference was also suggested from the magnetocaloric effect measurement [8]. Notice that T_1^{-1} gradually increases with increasing field above 14.5 T. In the plateau field region, T_1^{-1} decreases steeply with decreasing temperature [7], which presumably suggest the presence of an energy gap. Thus, we infer that low energy magnetic excitations are modified by the magnetic field while the spin structure keeps to be commensurate. It is noted here that the incommensurate phases below and above the plateau resemble each other from a viewpoint of NMR.

In summary, we have found a clear evidence of the first order phase transition between an incommensurate (cycloidal) and a commensurate (plateau) phases in ^{133}Cs-NMR spectrum and its relaxation rate T_1^{-1}. We also found an asymmetric behavior of T_1^{-1} within the plateau region. Detailed experimental results and discussion will appear elsewhere.

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